Fundamental Mechanisms, Predictive Modeling, and Novel Aerospace Applications of Plasma Assisted Combustion

Overview of OSU research plan

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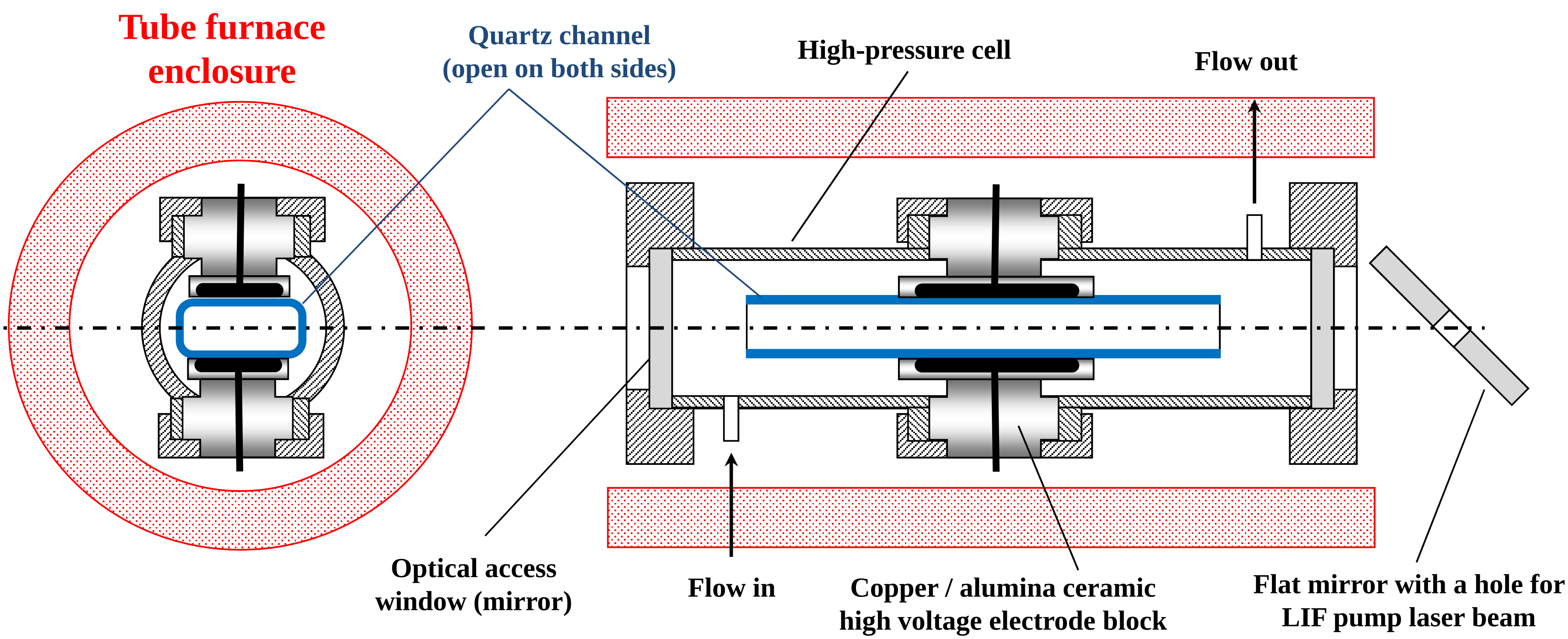
Thrust 1. Experimental studies of nonequilibrium air-fuel plasma kinetics using advanced non-intrusive diagnostics

Task 1: **Low-to-Moderate (T=300-800 K) temperature, spatial and time-dependent radical species concentration and temperature measurements in nanosecond pulse plasmas in a variety of fuel-air mixtures pressures (P=0.1 - 5 atm), and equivalence ratios (φ~0.1-3.0)**

Goal: **Generate an extensive set of experimental data on radical species concentrations and temperature rise; elucidate kinetic mechanisms of low-temperature plasma chemical fuel oxidation and ignition using kinetic modeling. Bridge the gap between room-temperature data (low-pressure gas discharges) and high-temperature data (shock tubes)**

Test Bed #1: High-temperature, high-pressure nanosecond pulse discharge cell



- High-pressure discharge cell inside a tube furnace (6 inch bore, up to $T=1200^0$ C)
- Premixed fuel-air flow (~1 m/s), preheated in the furnace, from 0.1 atm to a few atm
- Repetitive nanosecond pulse discharge plasma: 20-40 kV, 5-25 nsec, 10 Hz to 100 kHz
- Optical access (LIF, TALIF, CARS, CRDS) on the sides
- Fuels: hydrogen, methane, ethylene, propane, pentane, methanol & ethanol vapor

Repetitive nanosecond pulse plasma for kinetic studies:
Air, P=60 torr, ν=40 kHz, 40 msec burst, 1 μsec gate

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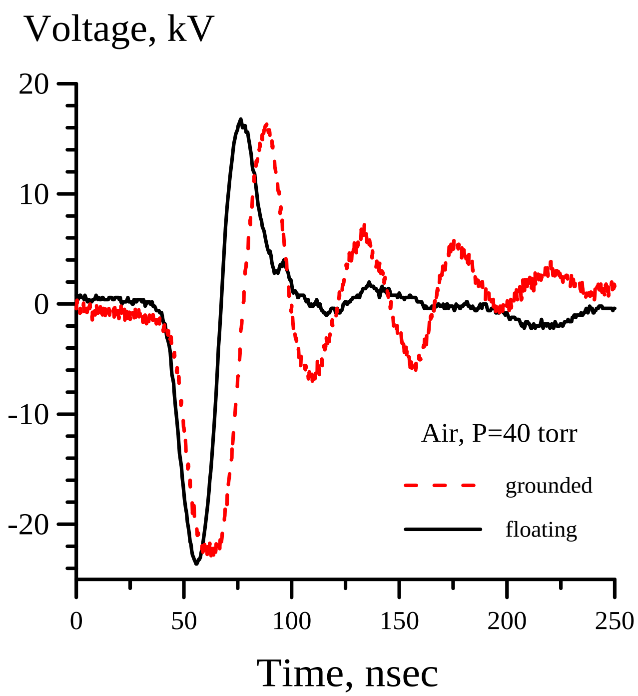
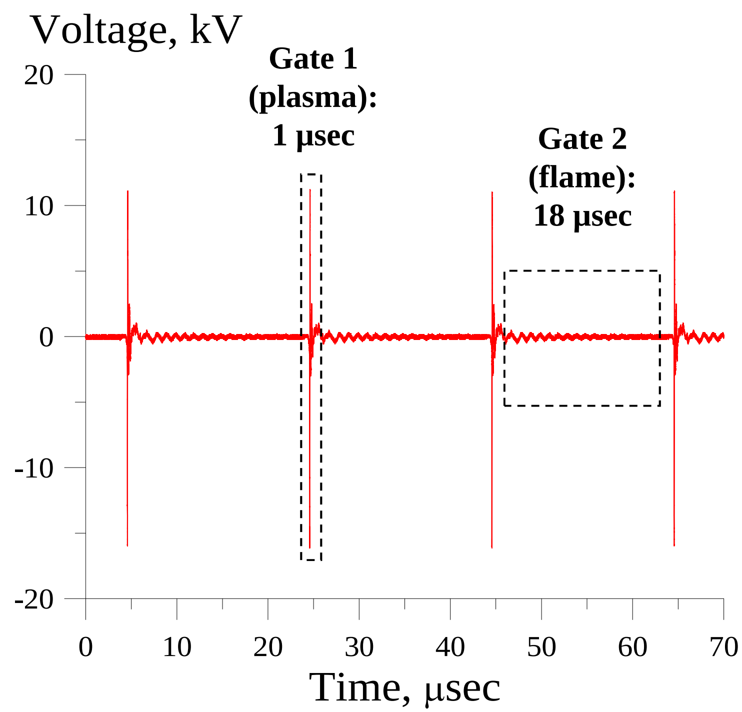
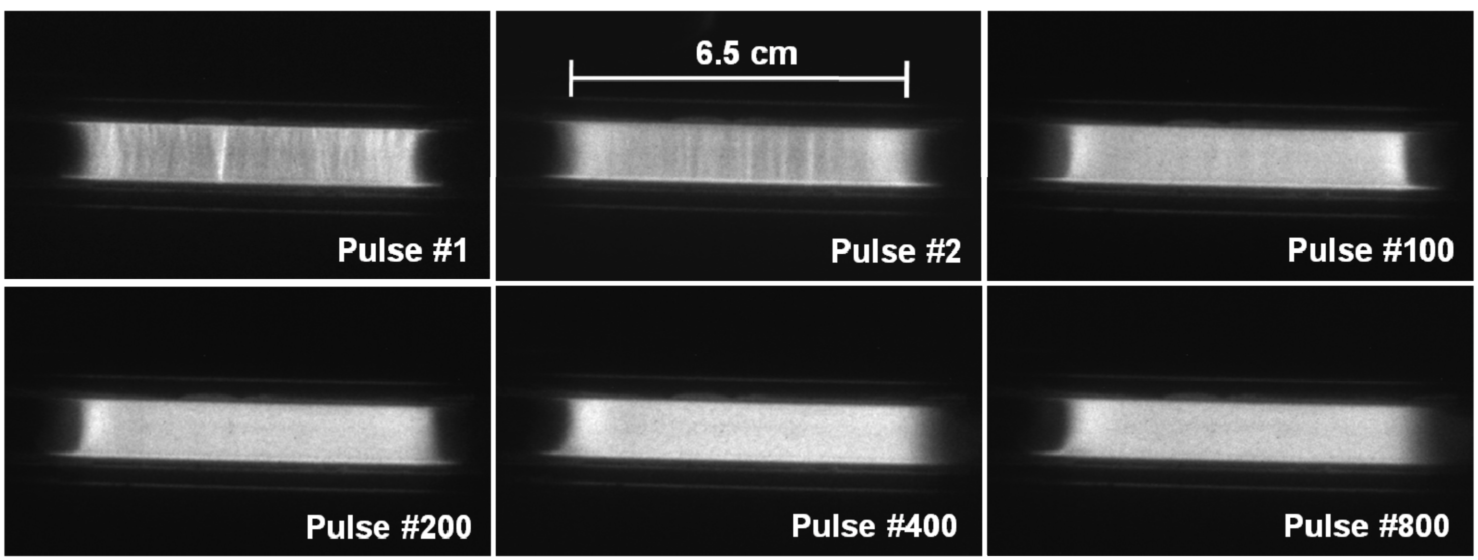


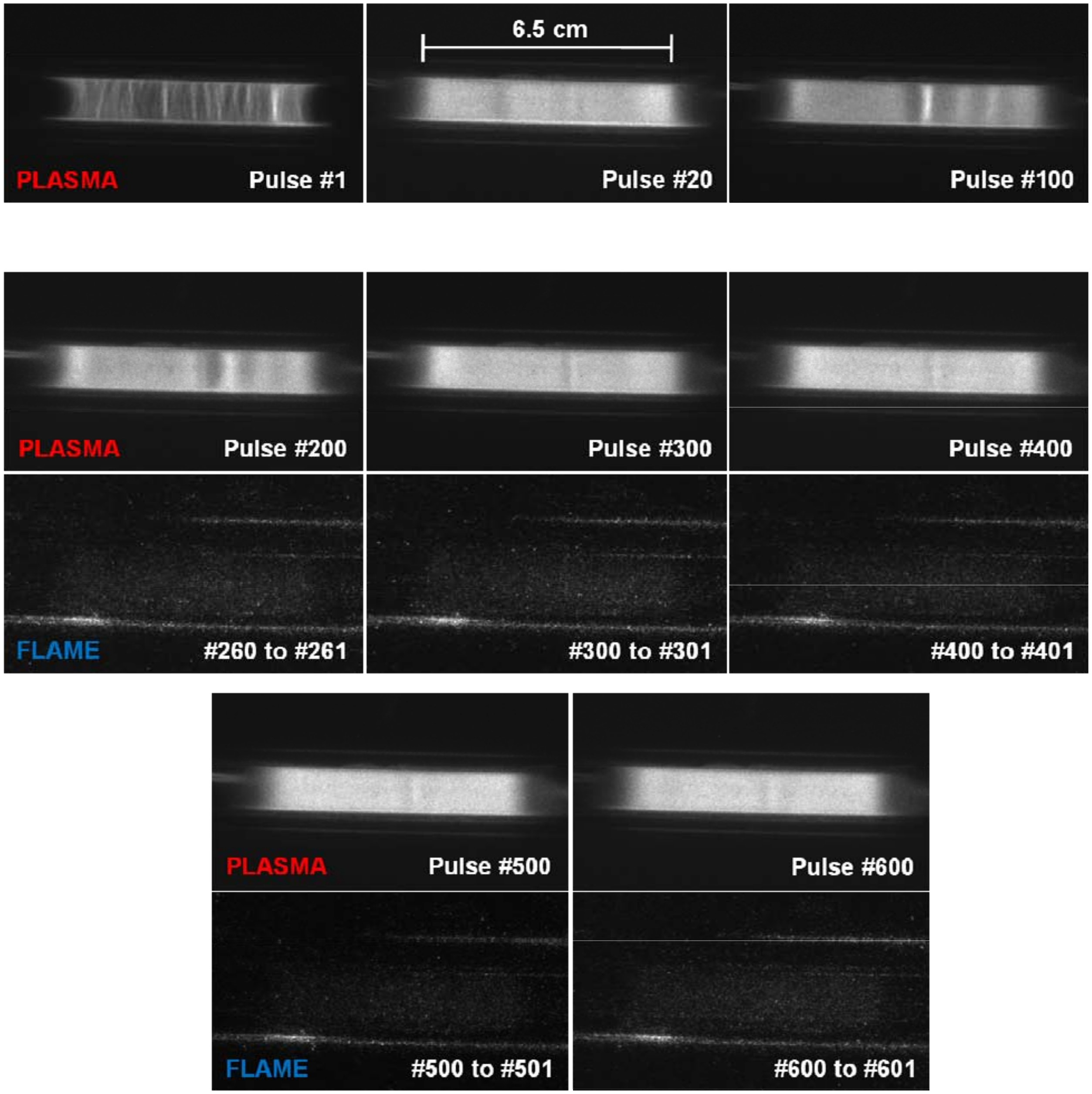


- **Some filamentary structure in pulses #1 and #2**
- **Uniform air plasma during subsequent pulses, at P=40-100 torr**



Repetitive nanosecond pulse plasma for kinetic studies:

Ethylene-air, P=40 torr, φ=1, ν=40 kHz



- Nearly uniform plasma during entire burst (except pulses #1 and #2)
- Ignition does not occur, likely due to rapid wall cooling
- Pressure is low – can this experiment be done at higher pressures?

Repetitive nanosecond pulse plasma for kinetic studies:
Ethylene-air, P=60 torr, φ=1, ν=40 kHz

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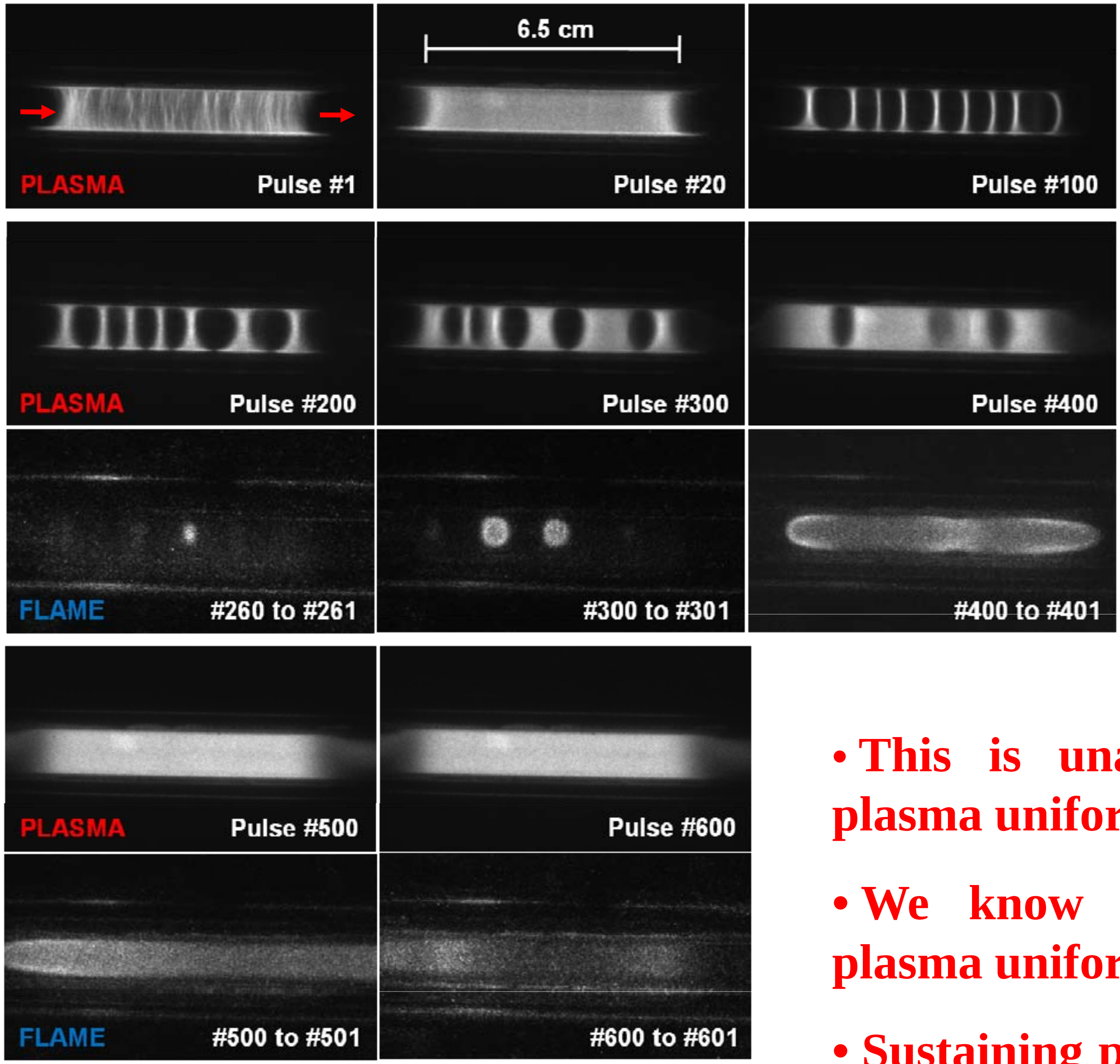

- Uniform plasma during first few tens of pulses (except pulses #1 and #2)
- Well-defined filaments form in pulse #100, persist for several hundred pulses
- After ignition occurs, flame fills entire discharge volume, and plasma becomes uniform again
- Filamentation likely due to ionization / heating instability

- This is unacceptable: need to keep the plasma uniform during entire burst
- We know that preheating will improve plasma uniformity
- Sustaining plasma in a heated cell will allow measurements at higher pressures

Time-resolved species concentrations: O and H atoms (Two-Photon Absorption LIF with Xe and Kr calibration)

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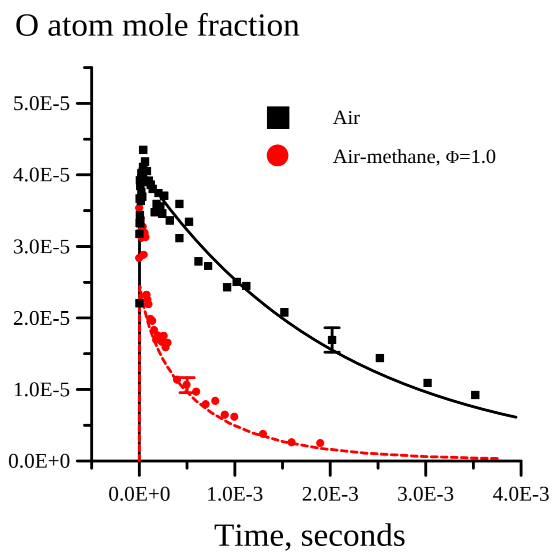

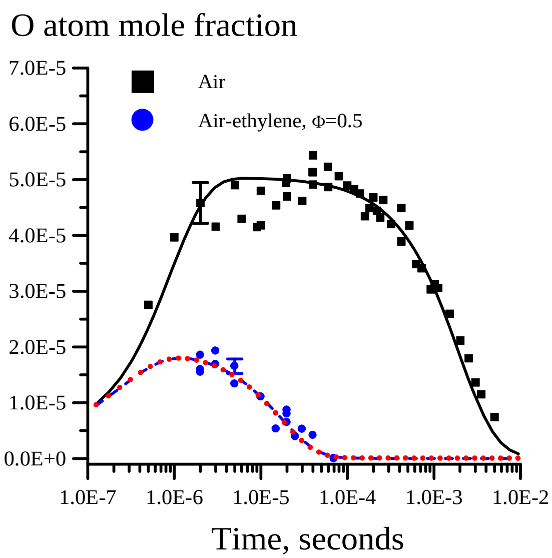

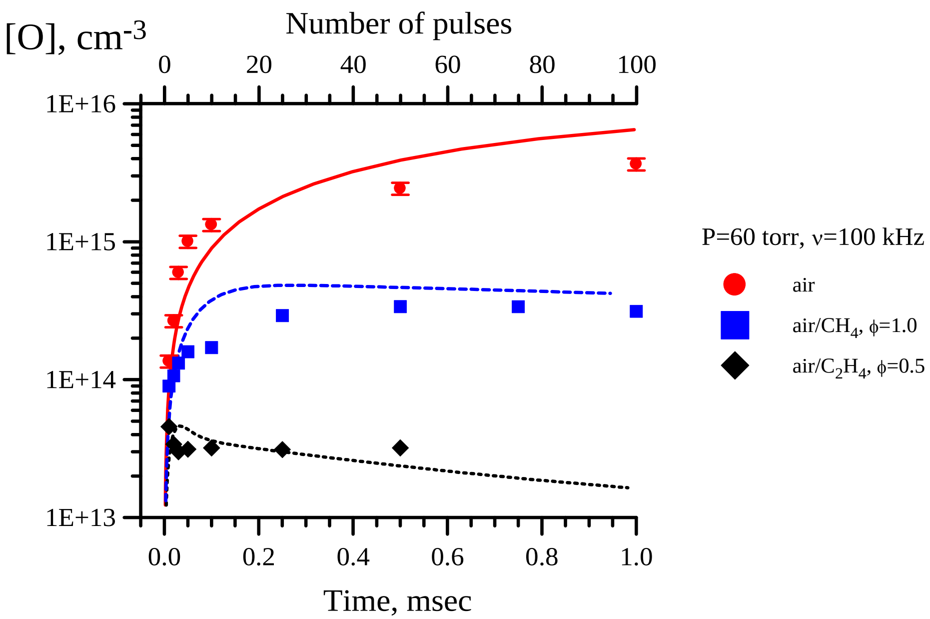

Previous results: O atoms in air, methane-air, and ethylene-air at P=60 torr (single-pulse and burst mode, initially at T=300 K)

Objective: measure time-resolved O and H atoms in nsec pulse discharge plasmas in H_2-air and C_xH_y air mixtures, at P ~ 0.1 - 1 atm, T=300-800 K

Outcome: kinetic mechanism of low-temperature plasma fuel dissociation and oxidation (specifically rates of O atom generation in the plasma and O atom reactions with fuel species)

Time-resolved species concentrations: OH
(LIF with Hencken adiabatic burner calibration)

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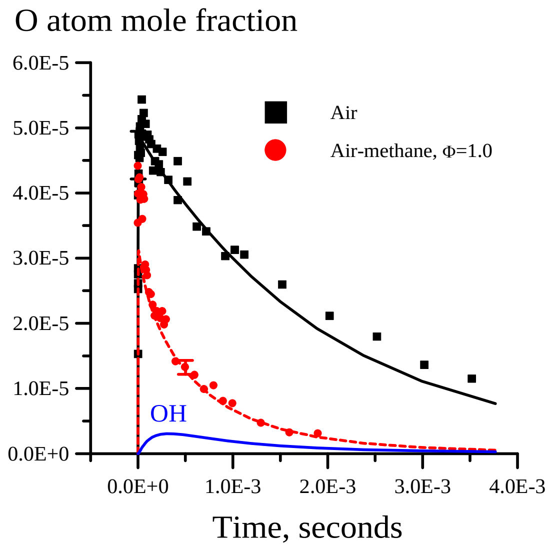
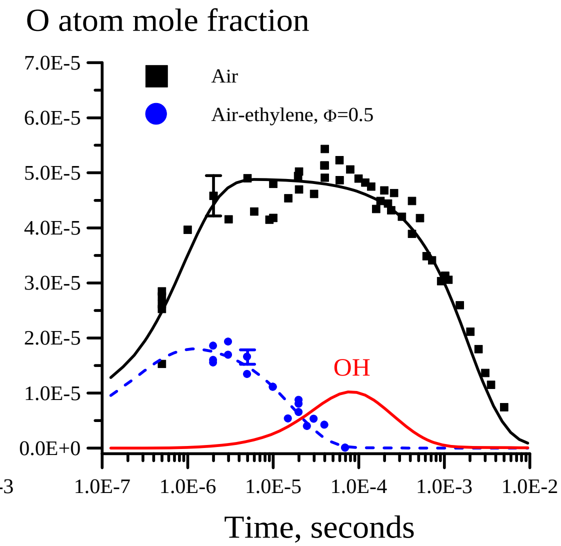
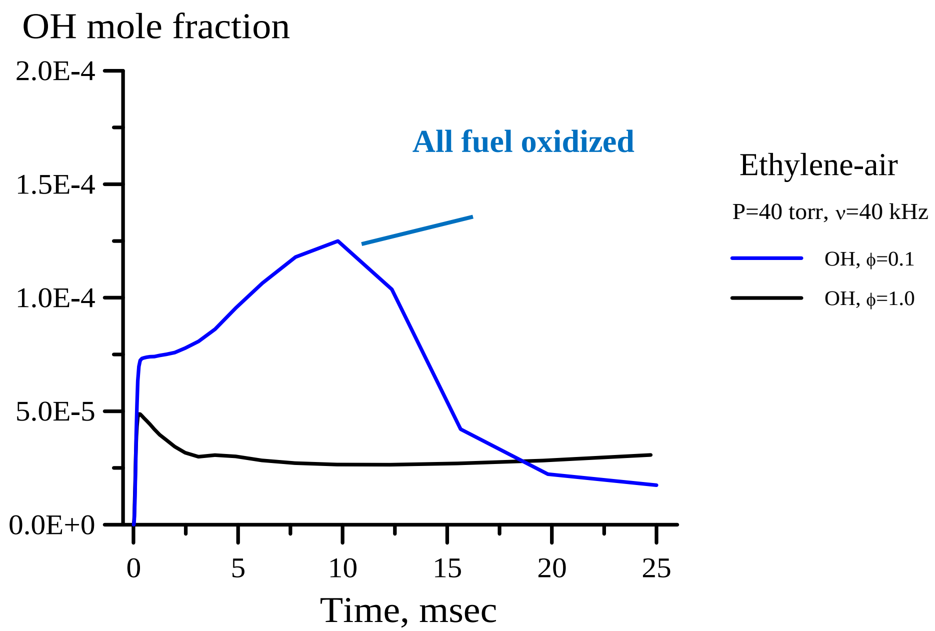




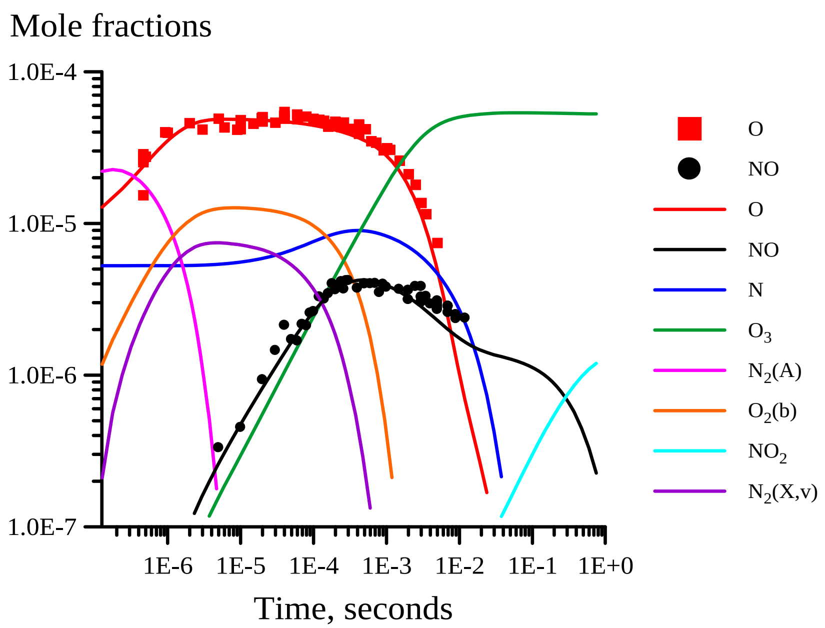


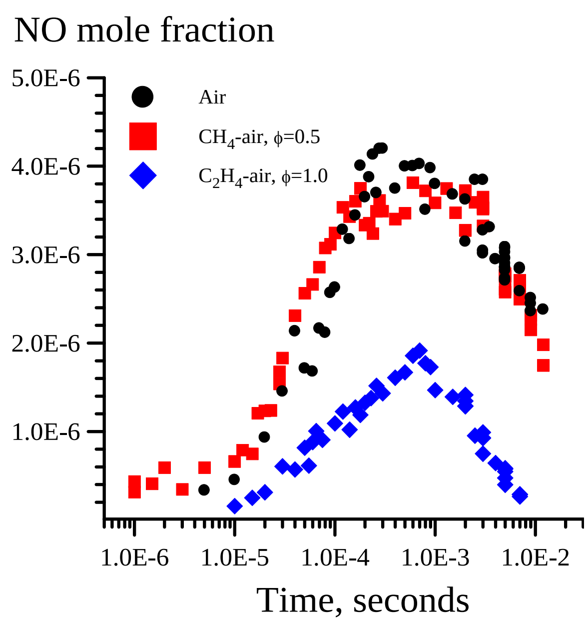
Work currently underway: OH in methane-air and ethylene-air at P=60 torr (single-pulse and burst mode, initially at T=300 K)

Objective: measure time-resolved OH in nsec pulse discharge plasmas in H_2-air and C_xH_y air mixtures, at P ~ 0.1 - 1 atm, T=300-800 K

Outcome: kinetic mechanism of low-temperature plasma fuel oxidation (specifically rates of H atom abstraction from fuel species)

Time-resolved species concentrations: NO
(LIF with calibration using known NO-N_2 mixture)



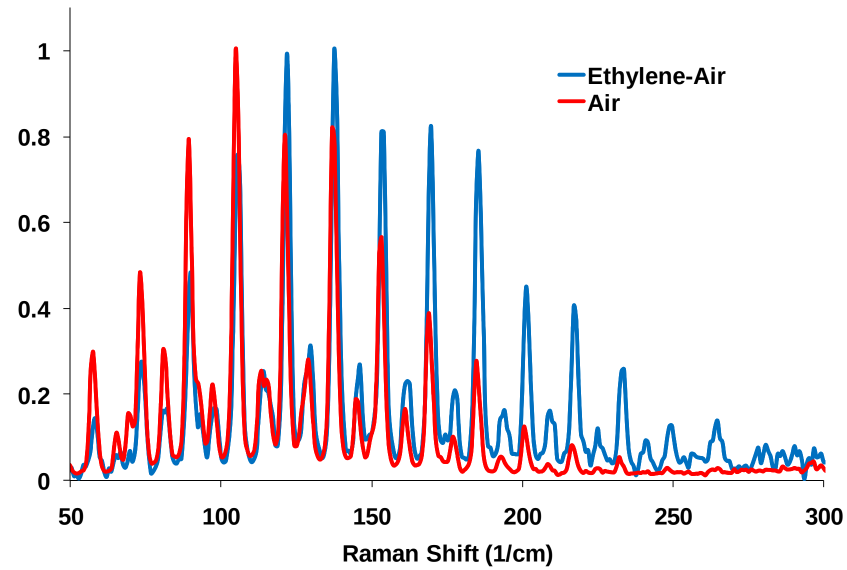


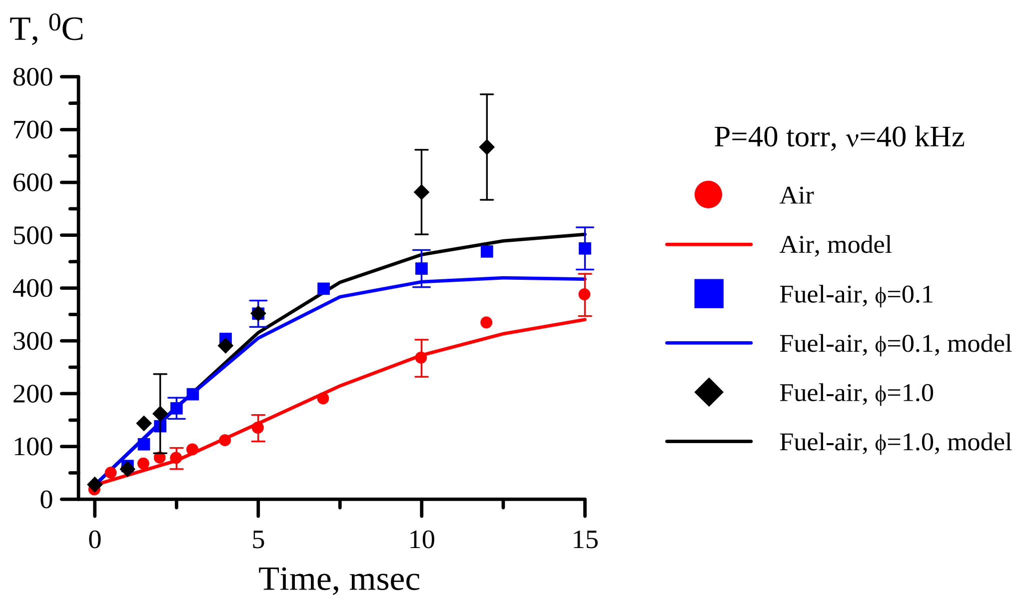
Previous results : NO in air, methane-air and ethylene-air at P=60 torr (single-pulse, initially at T=300 K). State-of-the-art kinetic models cannot explain time-resolved data. Possible effect of $N_2(X,v)$ + O reaction.

Objective: measure time-resolved NO in nsec pulse discharge plasmas in H_2-air and C_xH_y air mixtures, at P ~ 0.1 - 1 atm, T=300-800 K

Outcome: kinetic mechanism of low-temperature plasma fuel oxidation (specifically O_2 dissociation vs. NO formation in N_2* reactions)

Time-resolved, spatially resolved temperature (purely rotational CARS)



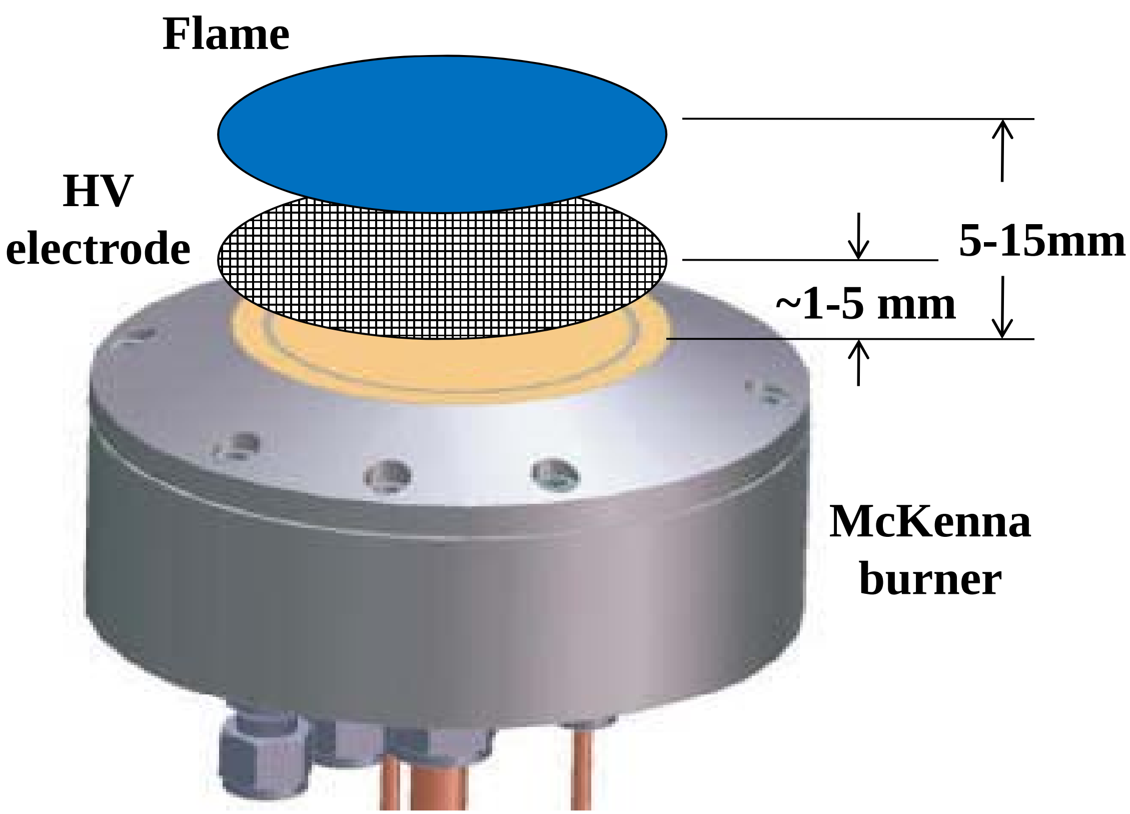


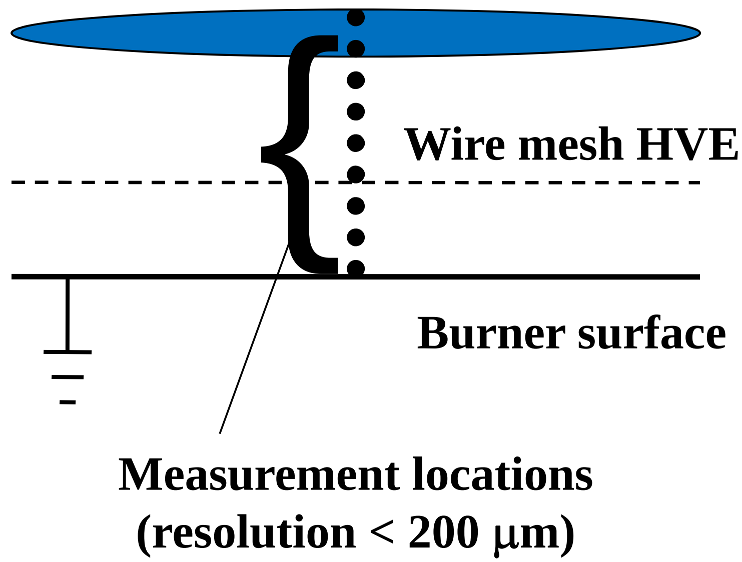
Previous results: time-resolved temperature in air and ethylene-air at P=40 torr (burst mode, initially at T=300 K). Evidence of significant additional heat release in fuel-air, compared to air

Objective: measure temperature in nsec pulse discharge plasmas in H_2-air and C_xH_y air mixtures, at P ~ 0.1 - 1 atm, T=300-800 K

Outcome: kinetic mechanism of low-temperature plasma chemical energy release in exothermic fuel oxidation reactions with radicals

Test Bed #2: Flat flame McKenna burner with nanosecond pulse discharge



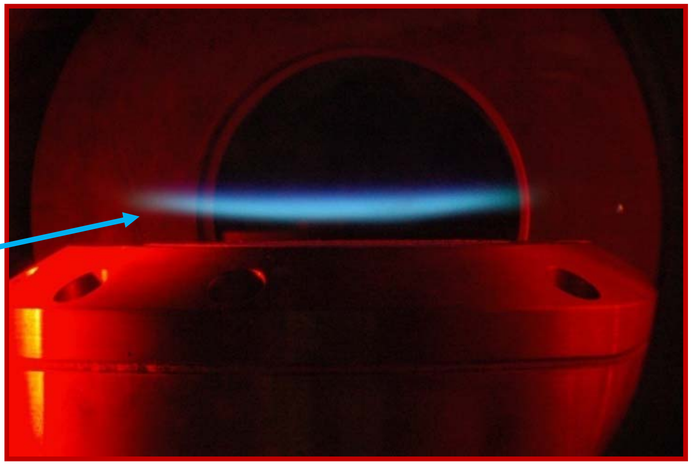


- Flat flame burner inside a six-arm cross vacuum chamber (8 inch bore)
- Premixed fuel-air flow (~0.1-1.0 m/s) with N_2 co-flow, P=10-40 torr
- Repetitive nanosecond pulse discharge plasma: 20-40 kV, 5-25 nsec, 10 Hz to 100 kHz
- Optical access (LIF, TALIF, CRDS) on two perpendicular axes
- Fuels: hydrogen, methane, ethylene, propane, pentane, methanol & ethanol vapor

Interaction of plasma and flame chemistry:
spatially resolved species concentrations and temperature

- **Steady, laminar, low-pressure flat flames allow spatially-resolved measurements of temperature and species concentrations**
- **Minimize transport influence; isolate kinetic effects**
- **Can investigate full range of temperature conditions (from below 500 K to 2000 K) by adjusting measurement position (i.e. height above burner)**
- **Typical spatial scale ~5-20 mm, spatial resolution <200 μm**
- **Straightforward integration of nsec discharge plasma into a low-pressure flame facility and study of plasma effects (i.e. measurements with plasma "off" and "on")**

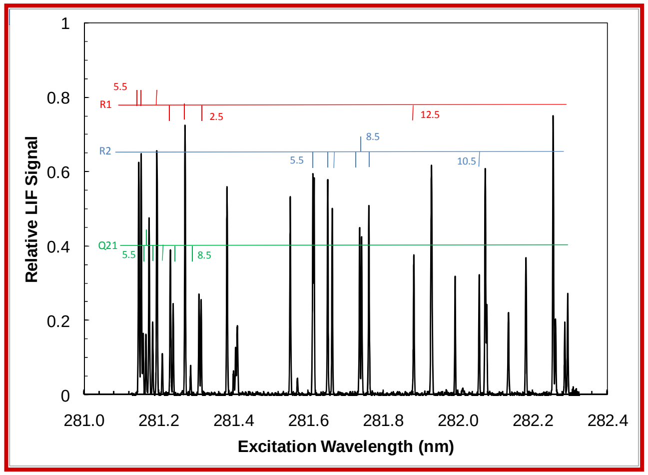
Steady, laminar, 30 Torr, 1-D flame

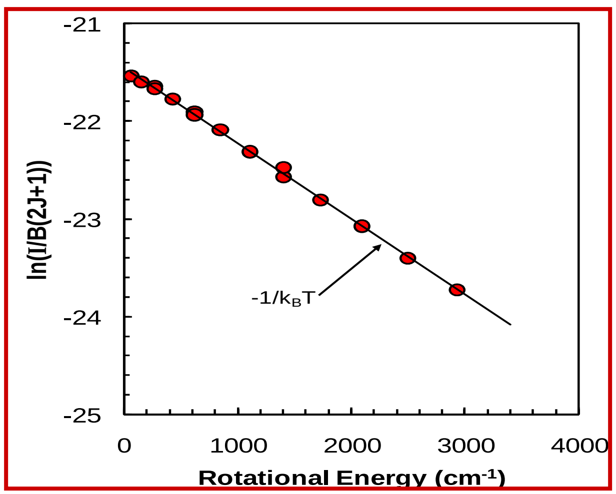


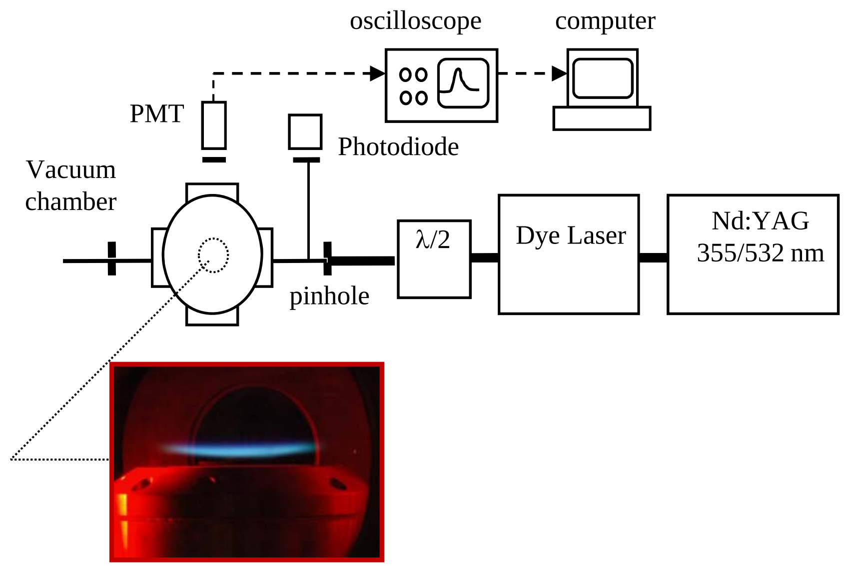
Previous low-pressure flame results (LIF):

P=10-40 torr; CH_4, C_2H_6, C_3H_8, C_4H_{10}; φ=0.6 -1.4

Flame temperature from rotational structure of **OH A-X (1,0)** band near 282 nm

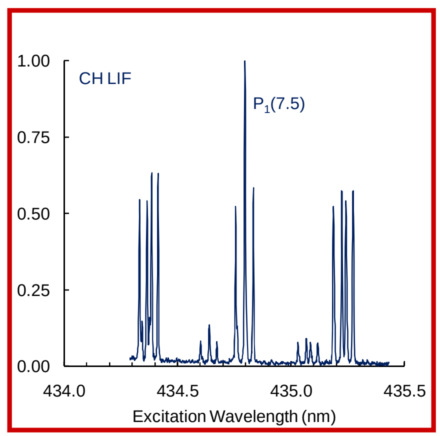


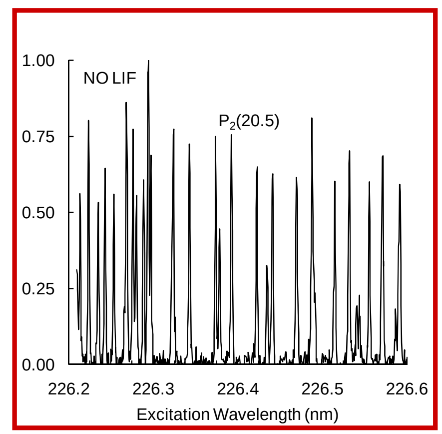




Spectral features used for profiles of flame species:

CH A-X (0,0) at 435 nm **NO A-X (0,0)** at 226 nm





Previous low-pressure flame results (LIF):
P=10-40 torr; CH_4, C_2H_6, C_3H_8, C_4H_{10}; φ=0.6 -1.4

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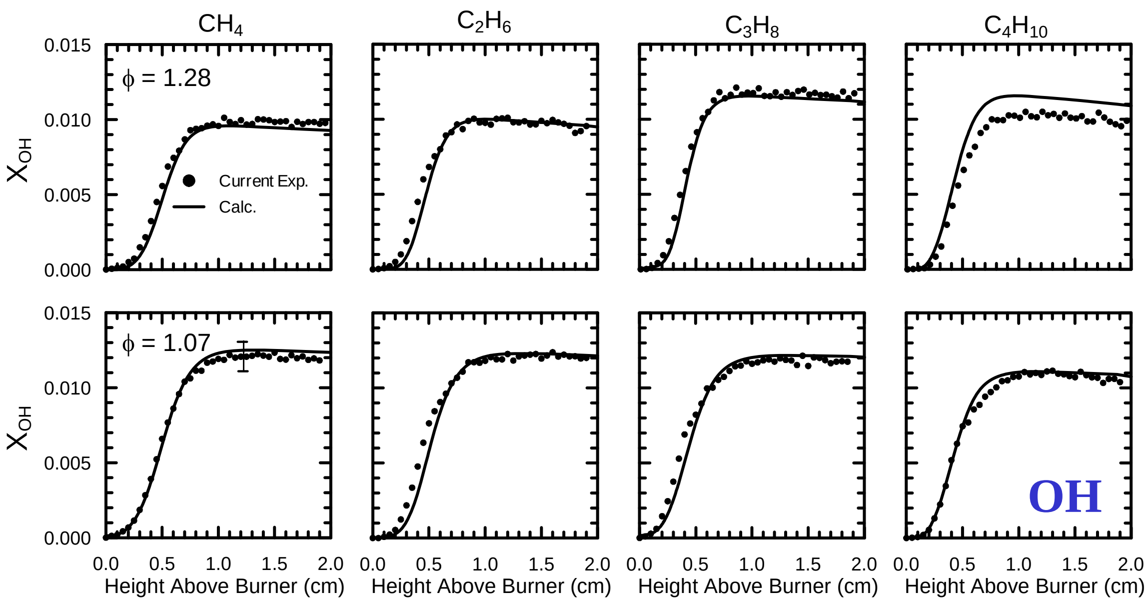
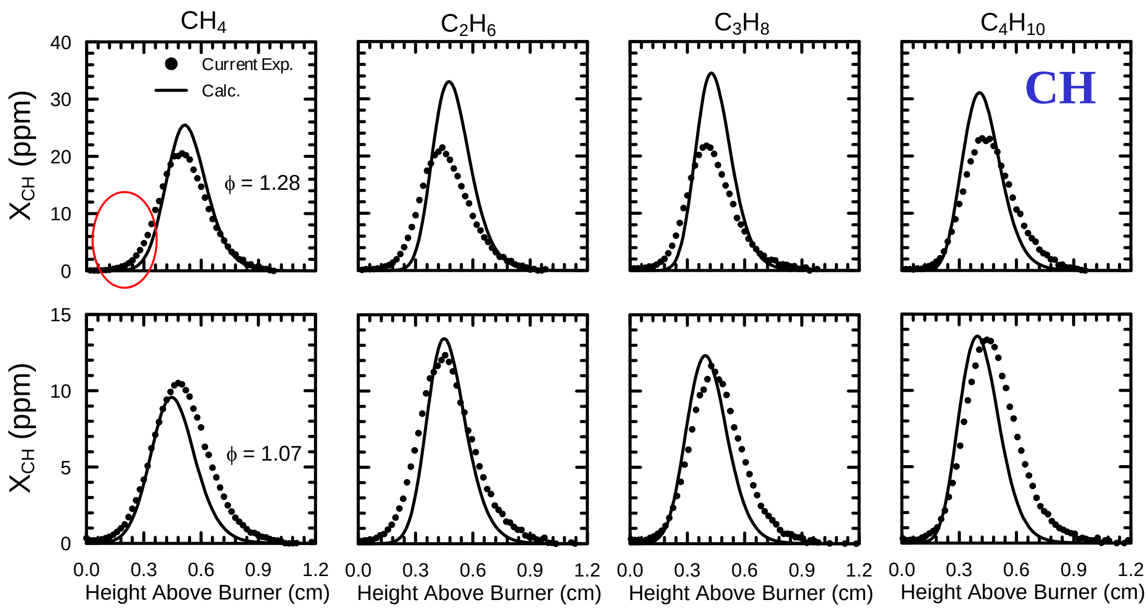


Spatially-resolved measurements of radicals to understand high-temperature flame chemistry, help kinetic model development

Kinetic modeling: GRI-Mech 3.0



We will look at the region upstream of the flame where coupling between plasma kinetics and flame chemistry is most important

Low-pressure flame / plasma measurements (LIF, CRDS)

Objective: Examine coupling of plasma and combustion kinetics in a 1-D low-pressure flame. Use spatially-resolved species concentration and temperature measurements by LIF (OH, H, O, and CH) and CRDS (HO_2, HCO, CH_3) to study the effect of quasi-steady (RF) and repetitively pulsed nsec discharge plasmas on low-temperature chemistry and coupling with the flame zone

Outcome: Kinetic mechanism of low-temperature plasma chemical fuel oxidation and energy release, and its effect on flame speed and burn rate. Specifically, boundary between "low-T" and "high-T" chemistry by measuring HO_2 radical concentration, at the conditions when O_2 is electronically excited

$$O_2 + H \rightarrow OH + O \qquad \text{(high temperatures)}$$

$$O_2 + H + M \rightarrow HO_2 + M \qquad \text{(low temperatures)}$$

CRDS diagnostics will be used in both "test bed" experiments, (I) high-T, high-P nsec discharge plasma cell, and (II) low-P flame / plasma cell

Thrust 2. Kinetic model development and validation

Task 8: **Development and validation of a predictive kinetic model of non-equilibrium plasma fuel oxidation and ignition, using experimental results of Thrust 1**

Goal: **Identify key mechanisms, reaction, and rates of plasma chemical fuel oxidation processes for a wide range of fuels, pressures, temperatures, and equivalence ratios. This is absolutely essential to predictive capability of the model.**

Current state of the art: hydrocarbon-air, low-temperature plasma chemistry kinetic model

- **Air plasma model: equations for ground state species (N, N_2, O, O_2, O_3, NO, NO_2, N_2O), charged species (electrons and ions), and excited species ($N_2(A^3\Sigma)$, $N_2(B^3\Pi)$, $N_2(C^3\Pi)$, $N_2(a'^1\Sigma)$, $O_2(a^1\Delta)$, $O_2(b^1\Sigma)$, $O_2(c^1\Sigma)$, $N(^2D)$, $N(^2P)$, $O(^1D)$) produced in the plasma.**
- **Two-term expansion Boltzmann equation for plasma electrons**
- **Fuel-air plasma: model combined with GRI Mech 3.0 C_xH_y oxidation mechanisms, supplemented with fuel dissociation by electron impact and in reactions with electronically excited nitrogen**
- **Peak E/N adjusted for pulse energy to be same as predicted by the nanosecond pulse discharge model**

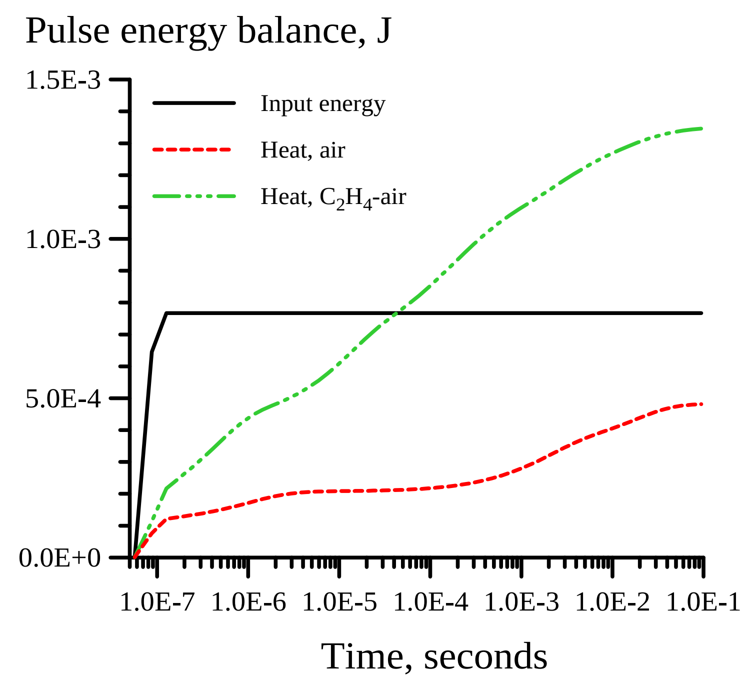
We have absolutely no reason to trust the model predictions: GRI Mech 3.0 (or any other combustion mechanism) is not designed to work at low temperatures (starting at T=300 K)

Confidence in the model can be provided only by detailed kinetic measurements such as discussed in Thrust 1 plan

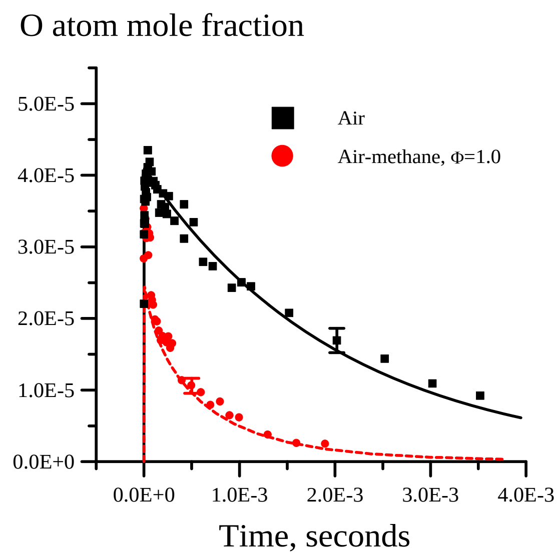
Here is what we know so far: dominant radical and energy release processes in C_2H_4-air predicted by the model

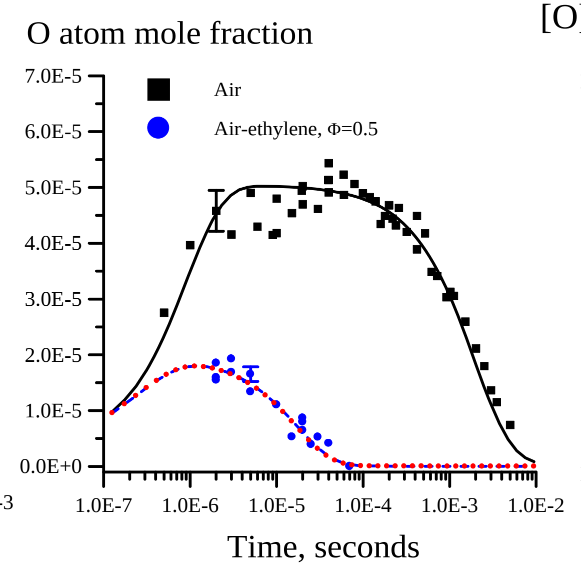
O atom generation
$N_2 + e^- = N_2(A^3\Sigma) + e^-$
$N_2 + e^- = N_2(B^3\Pi) + e^-$
$N_2 + e^- = N_2(C^3\Pi) + e^-$
$N_2 + e^- = N_2(a'^1\Sigma) + e^-$
$O_2 + e^- = O(^3P) + O(^3P,^1D) + e^-$
$N_2(C^3\Pi) + O_2 = N_2(a'^1\Sigma) + O_2$
$N_2(a'^1\Sigma) + O_2 = N_2(B^3\Pi) + O_2$
$N_2(B^3\Pi) + O_2 = N_2(A^3\Sigma) + O_2$
$N_2(A^3\Sigma) + O_2 = N_2 + O + O$
Fuel dissociation
$C_2H_4 + e\text{-} = \text{products} + e\text{-}$
$N_2(A^3\Sigma) + C_2H_4 = N_2 + C_2H_3 + H$
$N_2(B^3\Pi) + C_2H_4 = N_2 + C_2H_3 + H$
$N_2(C^3\Pi) + C_2H_4 = N_2 + C_2H_3 + H$
$N_2(a'^1\Sigma) + C_2H_4 = N_2 + C_2H_3 + H$
O atom decay
$O + C_2H_4 = CH_3 + HCO$
$O + C_2H_4 = H + CH_2CHO$
$C_2H_3 + O_2 = HCO + CH_2O$
$C_2H_3 + O_2 = O + CH_2CHO$
$O + O_2 + M = O_3 + M$
$O + O_3 = O_2 + O_2$

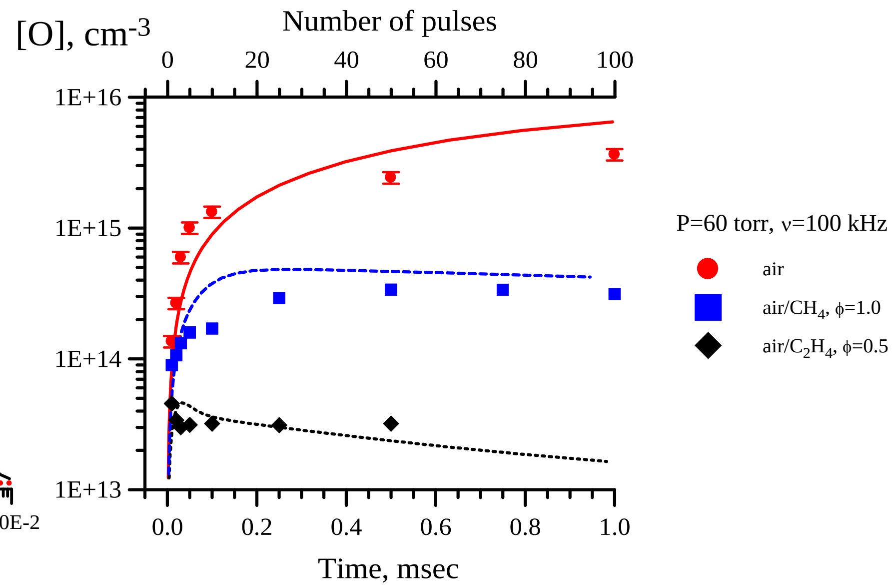
Fuel energy release
$O + CH_2CHO = H + CH_2 + CO_2$
$H + O_2 + M = HO_2 + M$
$O + HO_2 = OH + O_2$
$OH + HO_2 = O_2 + H_2O$
$OH + C_2H_4 = C_2H_3 + H_2O$
$HO_2 + CH_3 = OH + CH_3O$
$CH_3O + O_2 = HO_2 + CH_2O$
$O_2 + CH_2CHO = OH + HCO + HCO$
$HCO + O_2 = HO_2 + CO$
$HO_2 + HO_2 = O_2 + H_2O_2$
$CH_2 + O_2 = H + H + CO_2$

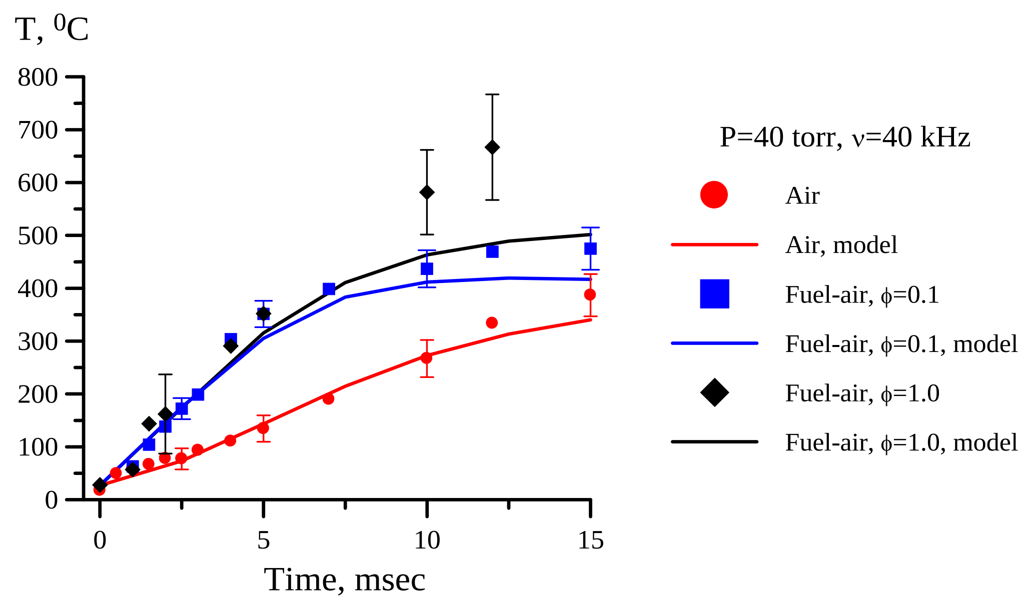


Model validation summary: so far so good…
… but no surprise if the model fails at some point









Need a lot more data from Thrust 1 for <u>extensive</u> model validation

Outcome: a self-consistent low-temperature fuel-air plasma chemical mechanism

Task 10: **Characterization and modeling of nsec pulse discharges**

Goal: **Prediction of E/N and electron density in the plasma, individual pulse energy coupled to the plasma, and their scaling with pressure, temperature, pulse waveform, and mixture composition**

Two-pronged approach to plasma assisted ignition modeling

Predictive modeling of energy release rate and ignition delay time in low-temperature, repetitive nanosecond pulse fuel-air plasmas requires:

- **E/N in the plasma, individual pulse energy coupled to the plasma, and their scaling with pressure, temperature, pulse waveform, and mixture composition**

- **Air plasma and fuel-air plasma chemistry: reactions among ground state species, excited species and radicals generated in the plasma, and their effect on energy release rate**

These two problems require separate analysis:

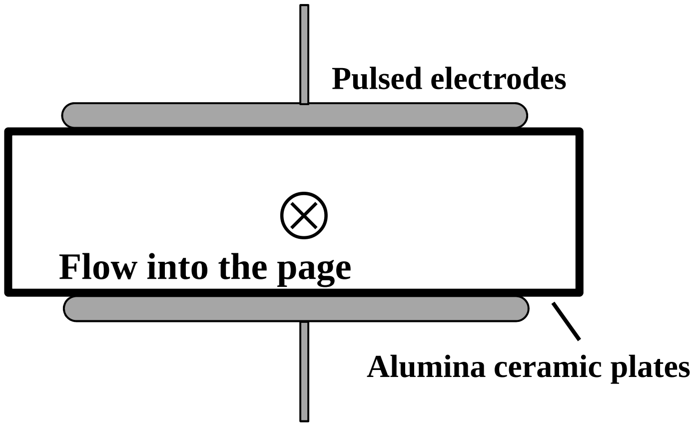
- **Nsec pulse plasma / sheath models cannot incorporate detailed reactive plasma chemistry: too many species (~100) and reactions (~1,000)**

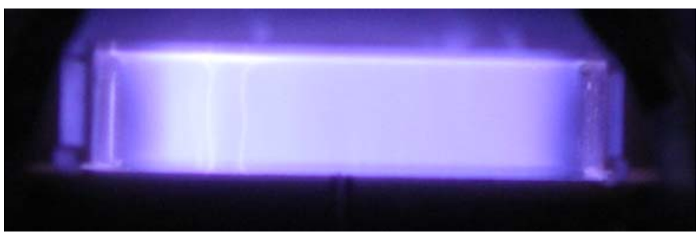
- **Detailed plasma chemistry models (quasi-neutral) cannot incorporate repetitive, nsec time scale sheath dynamics and plasma shielding**

Approach:

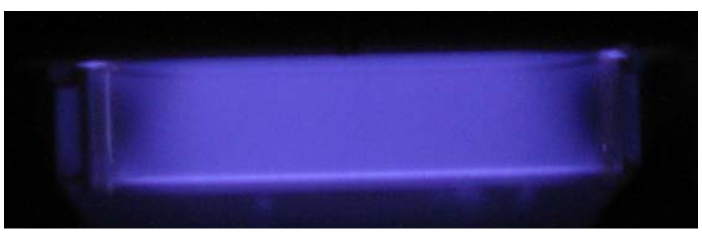
- **Predict plasma E/N and coupled pulse energy using nsec pulse plasma / sheath model**
- **Incorporate results into fuel-air plasma chemistry model**

Previous results:
Repetitive nsec discharge pulse energy measurements

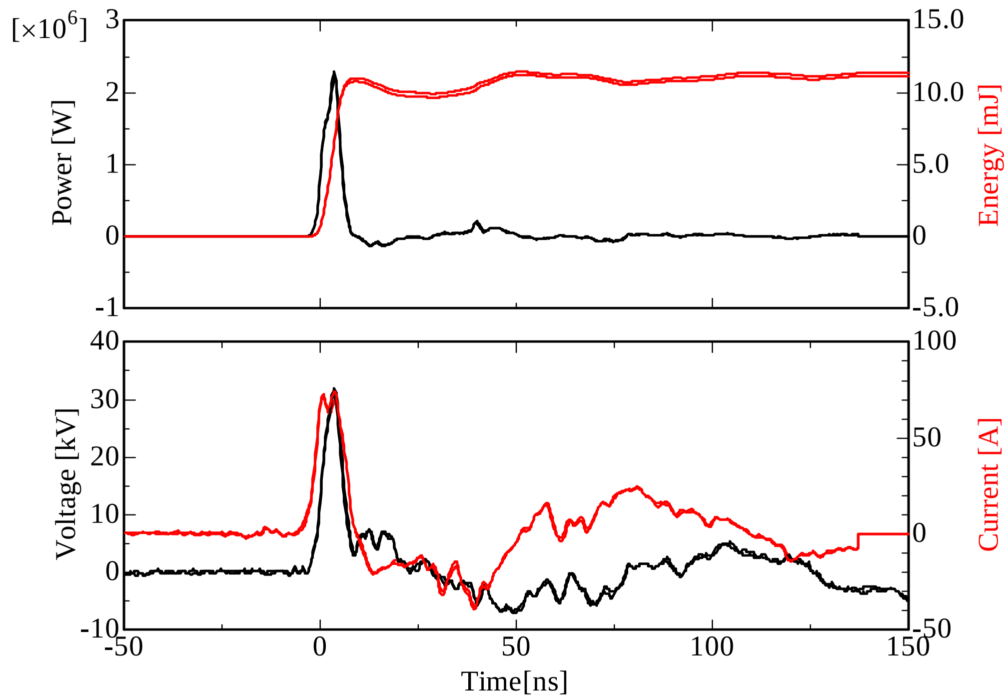




Nitrogen, P=300 torr, ν=100 kHz



Nitrogen, P=650 torr, ν=100 kHz



Nitrogen, P=350 torr, ν=100 kHz
0.3 seconds after start (pulse # 30,000)

Pulse energy 11 mJ/pulse
Discharge power 110 W

What are the electric field and the electron density?

Previous results:
Analytic nsec pulse discharge plasma / sheath model

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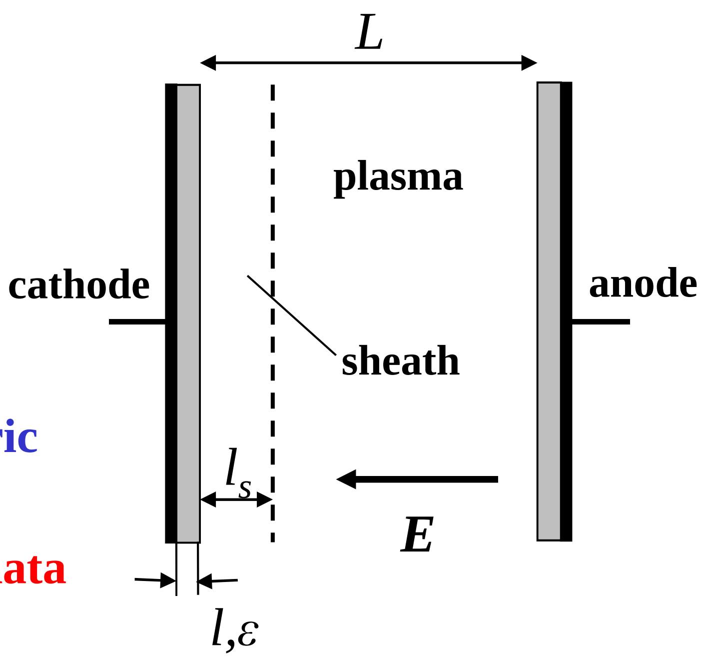
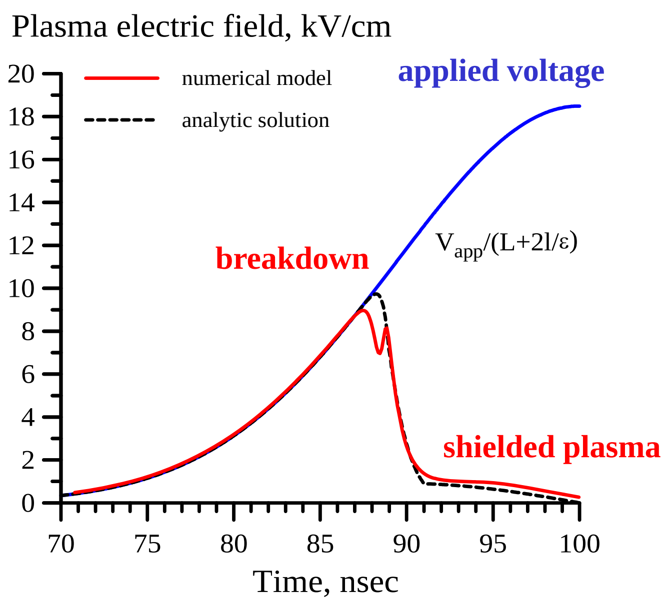
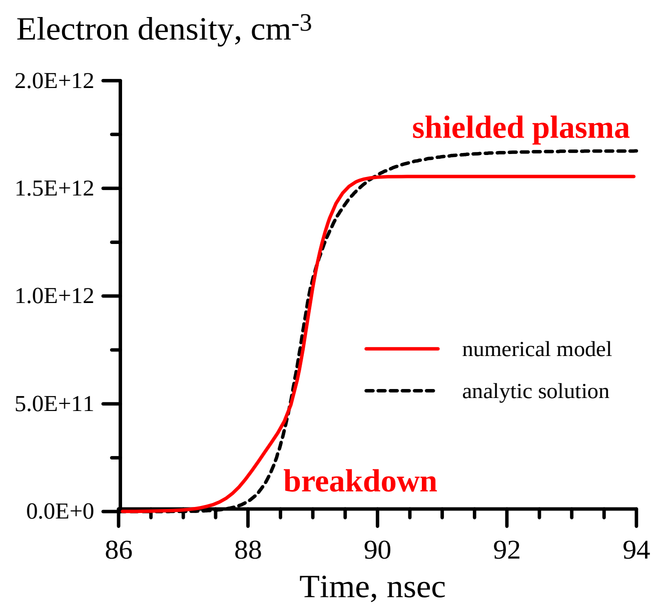
- **Equations for electron and ion number density**
- **Poisson equation for the electric field**
- **Plane-to-plane discharge geometry**
- **Voltage pulse: Gaussian fit to experimental waveform**
- **Dielectric plate charging / plasma shielding**



Analytic solution: time-dependent electron density and electric field in the plasma, coupled pulse energy

Excellent agreement with numerical solution, experimental data





Previous results:
Analytic nsec pulse discharge plasma / sheath model

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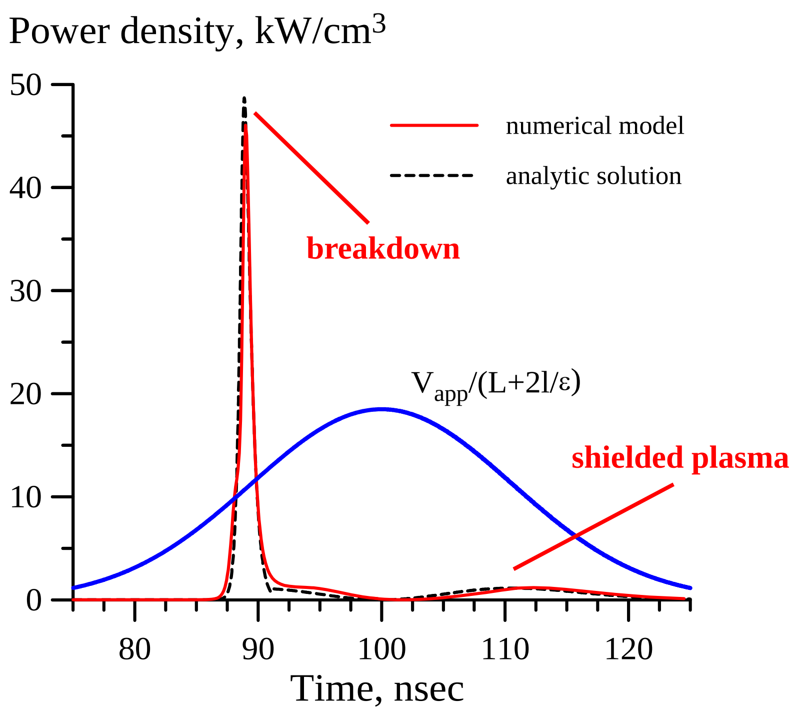
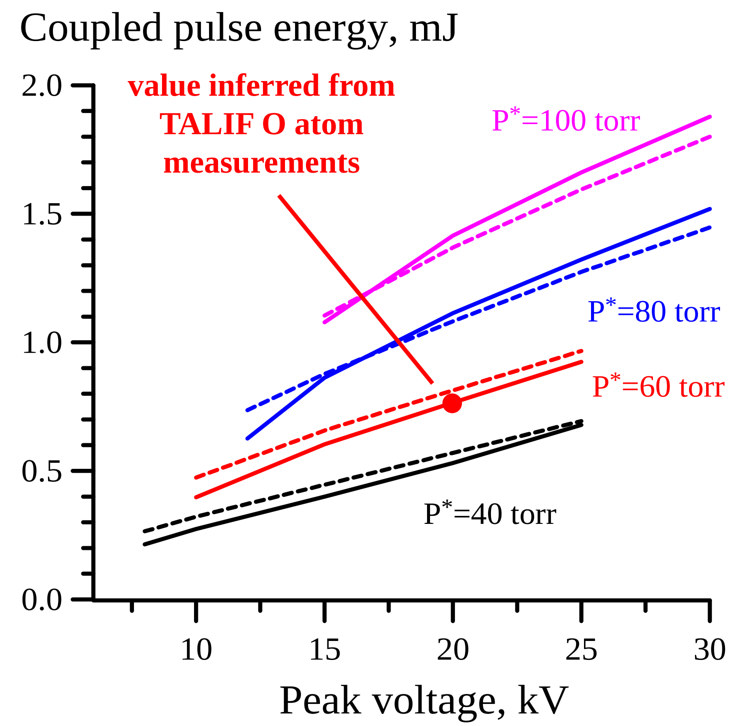


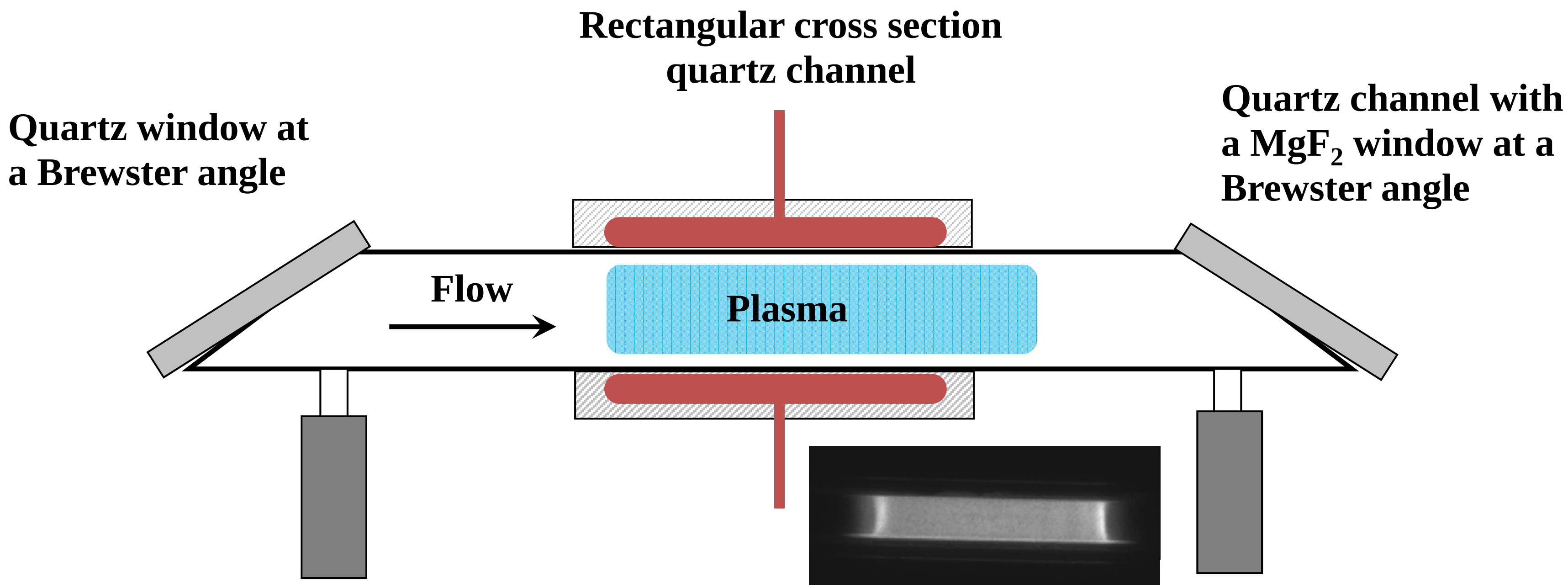


$$Q_{total} = Q_{break} + Q_{after} \approx \frac{1}{2} C_{load} V_{peak}^2 \left[\left(\frac{V_0}{V_{peak}} \right)^2 + \frac{\sqrt{2\pi}}{\nu_{RC} \tau_{pulse}} \right]$$

- **Coupled pulse energy scales with the number density, can be increased by increasing peak voltage, reducing pulse duration**

- **Excellent agreement with numerical solution, experimental data**

Electric field and electron density measurements: CARS, Thomson scattering



Entire test section mounted on a translation stage for spatially resolved measurements.

Objective: measure time- and space-resolved electric field and electron density in nsec pulse discharge plasmas using psec CARS and Thomson scattering; comparison with the model

Outcome: predictive capability for electron impact kinetic processes in the plasma

Task 12: **Ignition and flameholding in nonequilibrium plasma cavity flows at low static temperatures**

Goal: **Determine viable approaches to flameholding in high-speed flows using low-temperature plasmas. We simply cannot process the entire flow with the plasma!**

Previous results: cavity ignition in premixed ethylene-air flows by nsec plasma (25 kV, 20 nsec, 40 kHz)

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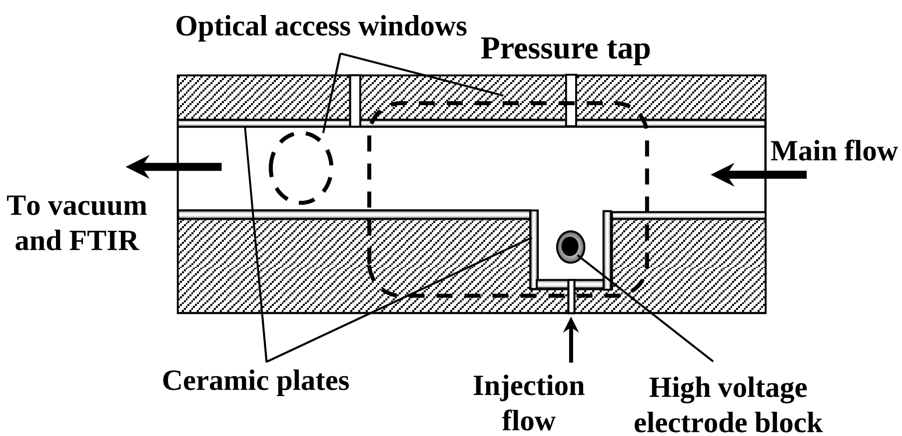
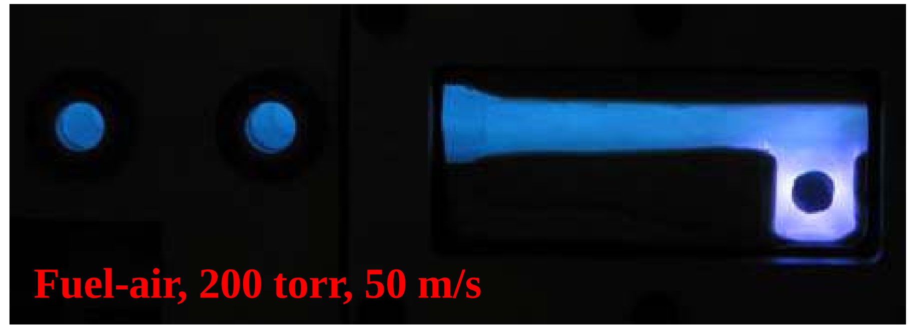
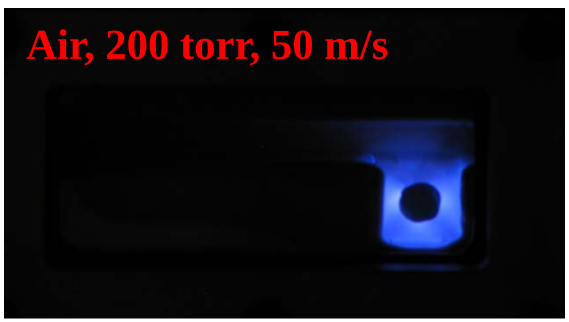
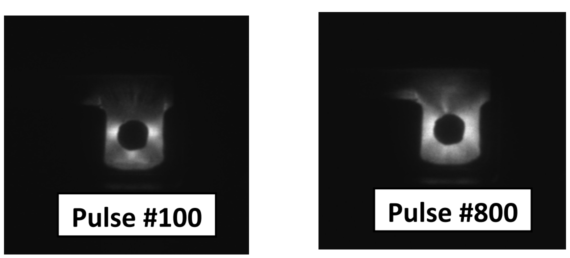
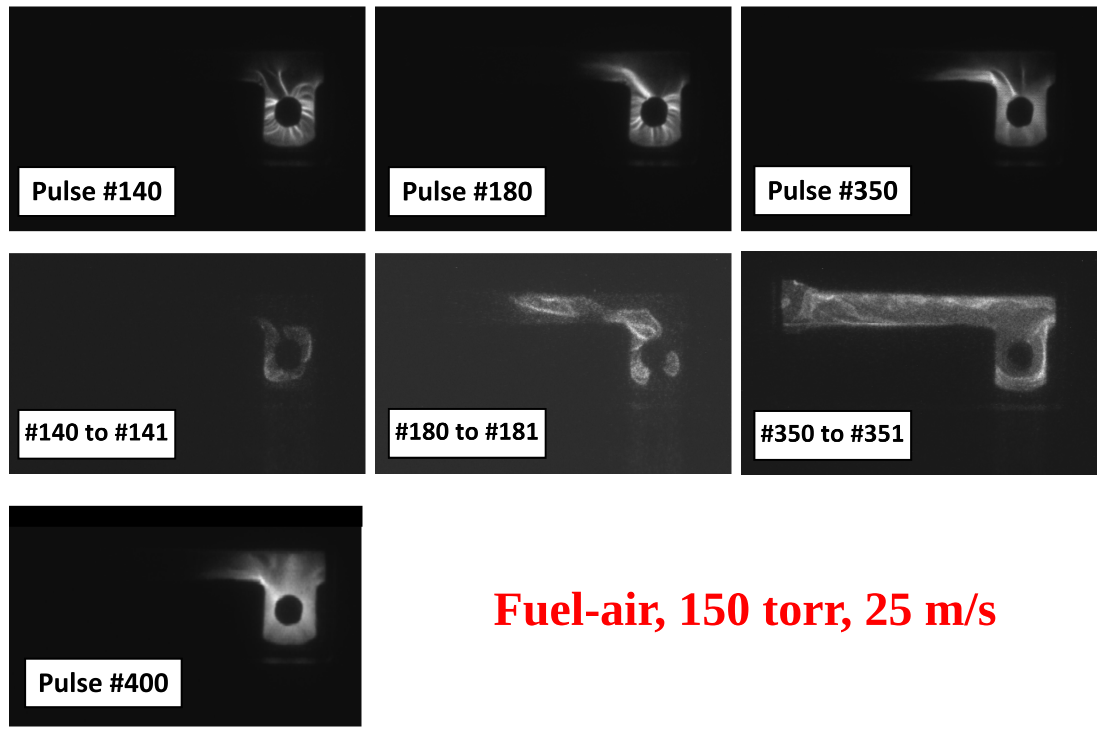








Air, 150 torr, 25 m/s



Fuel-air, 150 torr, 25 m/s

Diffuse plasma in air, filamentation in fuel-air during ignition, diffuse plasma after ignition

Previous results: cavity ignition and flameholding in premixed and non-premixed ethylene-air flows by nsec plasma

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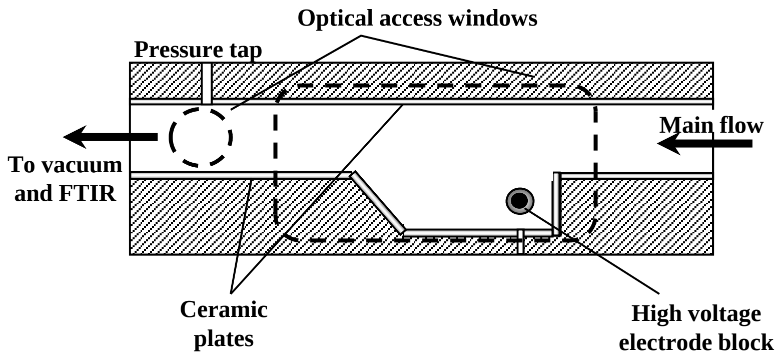
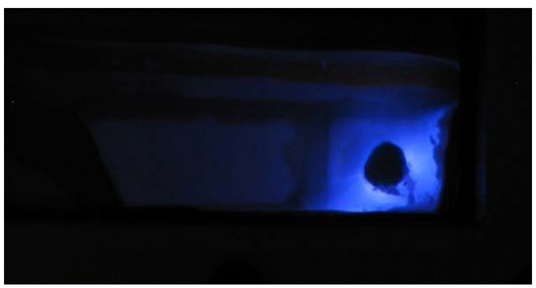
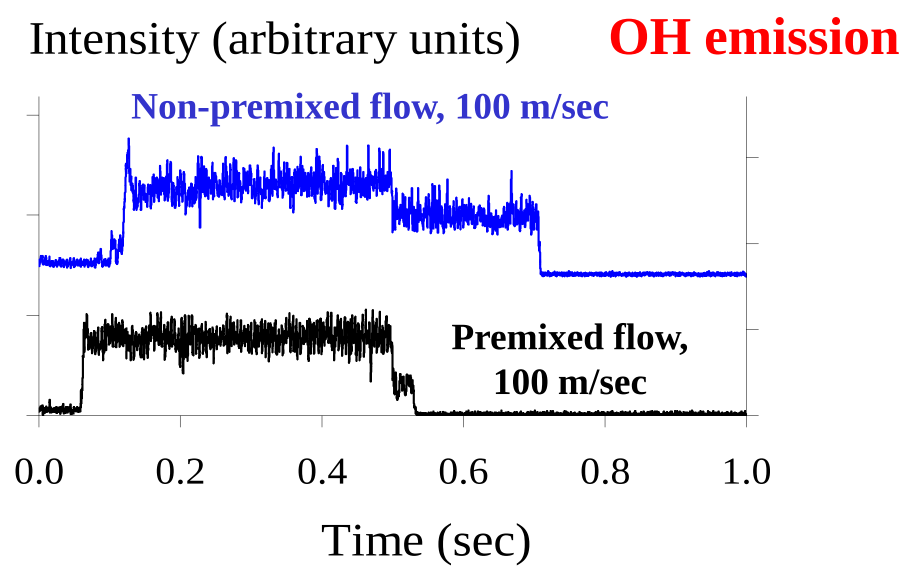




Fuel-air, 175 torr, 85 m/s



- Ignition and stable flameholding in both premixed and non-premixed flows up to 100 m/sec (global φ=1 in both cases)
- 80-90% burned fuel fraction
- Plasma power ~100 W, combustion energy release 35 kW
- After ignition, plasma needs to be "on" at all times (flame blow-off without plasma)

Ignition and flameholding in nonequilibrium plasma cavity flows

Objectives:

- **Further studies of cavity ignition and flameholding by repetitive nsec pulse plasmas in fuel injection flows (hydrogen and hydrocarbons)**
- **High frame rate (10-20 kHz) NO and OH PLIF imaging of ignition process using burst mode laser**
- **Increasing flow velocity beyond 100 m/sec, operating at low global equivalence ratios (φ=0.1-0.2)**
- **Comparison with kinetic modeling calculations using reduced plasma chemical ignition mechanism. Plasma flameholding mechanism after ignition – thermal or not?**

Outcome: **Demonstration of true predictive capability of the model**